Ejection Energy of Photoelectrons in Strong Field Ionization

D. Bauer
Theoretical Quantum Electronics (TQE), Technische Hochschule Darmstadt, Hochschulstr. 4A, D-64289 Darmstadt, Germany*

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We show that zero ejection energy of the photoelectrons is classically impossible for hydrogen-like ions, even when field ionization occurs adiabatically. To prove this we transform the basic equations to those describing two 2D anharmonic oscillators. The same method yields an alternative way to derive the anomalous critical field of hydrogen-like ions. The analytical results are confirmed and illustrated by numerical simulations.

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I. INTRODUCTION

A wealth of new phenomena in the interaction of strong, short laser pulses with matter has been found in recent years [1,2]. Laser systems delivering pulses at irradiances up to 10^{19} Wcm$^{-2}\mu$m^2 are available in several laboratories. Pulses not longer than a few tens of femtoseconds are "state of the art". In describing their interaction with matter usual perturbation theory fails since the electric field of such laser pulses is of the order of the atomic field. One of the prominent phenomena occuring in a strong laser pulse is field ionization. Here the electron is able to escape over the barrier formed by the Coulomb potential and the laser field. This process is much faster than tunneling which is dominant in weaker fields. In other words, in intense laser fields the so-called barrier suppression ionization (BSI) regime [3] is reached. In this regime, classical and semi-classical pictures are expected to work well and much theoretical research has been done in that direction, both analytically [4–6] and numerically [7–11].

So-called "simple man's theory" (SMT) [12–14] succeeded in explaining essential features of above threshold ionization (ATI) (see e.g. [1,15] for an overview), such as cut-offs in photoelectron energies and harmonic spectra [16]. Extensions to SMT considering rescattering effects clarified experimentally observed features such as plateaus in ATI-spectra [17] and rings in the angular distributions [18].

However, no ionization dynamics enters in the SMT since the electron is regarded as being "born" in the laser field without perceiving any attraction by the ionic core anymore, at least until rescattering by the nucleus. Even in Keldysh-Faisal-Reiss (KFR)-type theories [19–21] the Coulomb potential enters not directly but through the initial energy state wave function.

For relevant applications, e.g. inverse bremsstrahlung, it is important to know the energy with which the electron is freed (ejection energy) [22]. To calculate the latter the ionization dynamics must be described by solving the time-dependent Schrödinger equation or by using simpler alternative concepts. In this paper a classical model is used for this purpose.

For the sake of completeness, we briefly review SMT. Suppose that a linearly polarized laser field $E(t)$ sets in at $t = 0$, the velocity of a non-relativistic electron released at time t' is at the instant t

$$v(t) = -\int_{t'}^{t} E(\tau)\,\mathrm{d}\tau + v_{00} = -\int_{0}^{t} E(\tau)\,\mathrm{d}\tau + v_0(t') + v_{00} \tag{1}$$

(atomic units will be used throughout this paper) with

$$v_0(t') = -\int_{t'}^{0} E(\tau)\,\mathrm{d}\tau$$

the residual drift due to ejection out of phase, i.e. *not* in the maxima of the field. v_{00} is the ejection velocity we are particularly interested in. Since the ejection velocity v_{00} contributes to the overall residual kinetic energy $\mathcal{E}_{\rm res}$ of the electron at the end of the pulse,

$$\mathcal{E}_{\rm res} = \frac{1}{2}m(v_0 + v_{00})^2,$$

it alters the electron distribution function and must be taken into account. Ignoring v_{00} in (1) leads in the case of linear polarized laser light to the well-known results $\langle\mathcal{E}_{\rm res}\rangle = 3U_p$, $\max\mathcal{E}_{\rm res} = 8U_p$ as well as to the maximum return energy $\approx 3.2U_p$ which is responsible for the cutoff in harmonic-spectra. U_p is the ponderomotive potential $e^2E^2/(4m\omega^2)$, i.e. the mean quiver energy of the electron in the field.

The paper is organized as follows: in Sec. II the anharmonic oscillator picture is introduced which leads to a physically appealing interpretation of the adiabatic ionization process as discussed in Sec. III. In Sec. IV, the lower limit for the ejection energy as well as the upper limit for the ejection radius is derived. It is further shown that no upper limit for the ejection energy exists. In Sec. V the analytical results are confirmed and illustrated by numerical simulations. Sec. VI is devoted to a rederivation of the anomalous critical field of hydrogen like ions [26] in the framework of the anharmonic oscillator picture. The adiabatic deformation of the initial ground state is calculated and compared with the numerical solution of the time-dependent Schrödinger equation. Finally, we give our conclusion in Sec. VII.

II. TRANSFORMING TO ANHARMONIC OSCILLATORS

The laser frequency Ω can be regarded as small compared to the orbital frequency ω of the bound electron (e.g. $\Omega = 0.18$ a.u. for KrF and $\Omega = 0.04$ a.u. for Nd). Therefore, a quasi adiabatic treatment is appropriate. We will now outline the transformation to a set of two 2D anharmonic oscillators [23].

We start with the Hamiltonian describing an electron in a Coulomb potential $-Z/\sqrt{\rho^2+z^2}$, and a static electric field E aligned in z direction,

$$H(\rho, z; p_\rho, p_z, p_\varphi) = \frac{1}{2}\left(p_z^2 + p_\rho^2 + \frac{p_\varphi^2}{\rho^2}\right) - \frac{Z}{\sqrt{\rho^2+z^2}} + Ez = \mathcal{E}. \tag{2}$$

Here, $\mathcal{E}$ is the total energy of this conservative system and the azimuthal momentum p_φ is a constant of the motion.

We transform to parabolic coordinates first. The new coordinates ξ and η are related to the cylindric ones according to

$$\xi = (r - z)/2, \quad \eta = (r + z)/2,$$

with $r = \sqrt{\rho^2+z^2}$. The canonical momenta are related through

$$p_\xi = \sqrt{\frac{\eta}{\xi}}p_\rho - p_z, \quad p_\eta = \sqrt{\frac{\xi}{\eta}}p_\rho + p_z.$$

This leads to the Hamiltonian in parabolic coordinates,

$$H(\xi, \eta; p_\xi, p_\eta, p_\varphi) = \frac{1}{2}\left(\frac{\xi}{\xi+\eta}p_\xi^2 + \frac{\eta}{\xi+\eta}p_\eta^2 + \frac{1}{4\xi\eta}p_\varphi^2\right) - \frac{Z}{\xi+\eta} + E(\eta - \xi) = \mathcal{E}. \tag{3}$$

It is well known that the Hamilton-Jacobi-Equation of the problem separates in parabolic coordinates [24], as does so Schrödinger's equation. It is advantageous to perform another canonical transformation [23],

$$\xi = u^2/4, \quad \eta = v^2/4, \quad 0 \le u, v < \infty,$$

$$p_u = \sqrt{\xi}p_\xi, \quad p_v = \sqrt{\eta}p_\eta,$$

leading to

$$H(u, v; p_u, p_v, p_\varphi) = \frac{1}{2}\left(\frac{4}{u^2+v^2}\right)\left(p_u^2 + \frac{1}{u^2}p_\varphi^2 + p_v^2 + \frac{1}{v^2}p_\varphi^2\right) - \frac{4Z}{u^2+v^2} + g(v^2 - u^2) = \mathcal{E}, \tag{4}$$

where the new field $g = E/4$ has been introduced.

Now, we define the "zero energy Hamiltonian" $H_0 = H - \mathcal{E} \equiv 0$, and by multiplying this equation with $(u^2+v^2)/4 = r$ we are left with

$$H_0' = \frac{1}{2}\left(p_u^2 + \frac{p_\varphi^2}{u^2} + p_v^2 + \frac{p_\varphi^2}{v^2}\right) + \frac{\omega^2}{2}(u^2+v^2) - Z + \frac{1}{4}g(v^4-u^4) \equiv 0,$$

where $\omega^2 = -\mathcal{E}/2$. Therefore, we finally get

$$H_u(u; p_u, p_\varphi) = \frac{1}{2}\left(p_u^2 + \frac{p_\varphi^2}{u^2}\right) + \frac{\omega^2}{2}u^2 - \frac{1}{4}gu^4 = A, \tag{5}$$

$$H_v(v; p_v, p_{\varphi'}) = \frac{1}{2}\left(p_v^2 + \frac{p_{\varphi'}^2}{v^2}\right) + \frac{\omega^2}{2}v^2 + \frac{1}{4}gv^4 = B, \tag{6}$$

$$A + B = Z, \tag{7}$$

$$p_\varphi = p_{\varphi'}, \tag{8}$$

which represents a set of two 2D anharmonic oscillators moving independently. There are only constraints concerning the *initial* values of the system: (i) the energies must sum up to the given total "energy" Z, and (ii) the angular momenta, which are constants of the motion, are equal.

III. PHYSICAL PICTURE

The physical interpretation of Eqs.(5) and (6) is very simple. Supposing $g \geq 0$, Eq.(6) allows only for a bound motion in v, whereas the effective potential in Eq.(5), $V_u(u) = p_\varphi^2/(2u^2) + \omega^2 u^2/2 - gu^4/4$, has a local maximum so that the motion in u is unbound if the energy A lies above the potential barrier.

Now, the field g may be turned on adiabatically. Initially, when $g = 0$ holds, the v- and the u-motion take place in equal potentials. While g increases, the u-potential is bent down whereas the v-potential is steepened. Therefore one expects an adiabatic lowering of the energy A and an adiabatic raising of the level B (Fig. 1). But the frequency ω may also change because the adiabatic invariant

$$S_u(\omega, g, A) = \sqrt{2}\oint \sqrt{A - V_u(u)}\,\mathrm{d}u = \text{const.} \tag{9}$$

yields a relation between g, A *and* ω only. Indeed, a change in ω is the classical dc Stark-shift, of course. The trick is to consider the special Kepler orbit which ionizes earliest in an adiabatically ramped dc field. Therefore, we switch back to the "physical space" for a moment: the classical trajectory which ionizes first is that one mostly directed towards the potential barrier. There are two Kepler orbits lying on the z-axis in the *limit* eccentricity $\to 1$. One lies in the region $z \geq 0$, the other one in $z \leq 0$. If the potential barrier comes from negative z, as in our case where $g \geq 0$, the Kepler orbit $z \leq 0$ ionizes first and the Kepler orbit $z \geq 0$ does last. The two z-directed orbits are the extreme Stark-shifted ones. The orbit which ionizes first corresponds to the electron with smallest ejection energy. Therefore, the ejection energy of just this electron provides a lower limit for all ejection energies which may occur. Fortunately, this special energy can be calculated.

Now we turn back to the anharmonic oscillator picture: motion along the negative z-axis means $v = p_\varphi = p_{\varphi'} = 0$ for all times. Therefore, the anharmonic oscillator H_v is frozen, i.e., $B \equiv 0$. Now, when g is adiabatically ramped B is *not* adiabatically raised since the v-particle *rests* in the potential well and does not recognize the steepening of the potential. It follows that B remains zero, and $A \equiv Z$ (Fig. 2). The adiabatic invariant (9) now provides a relation between ω and g only, i.e., one can, in principal, calculate the classical Stark-shift $\omega^2(g)$ for this particular orbit. The Stark-shift is the key which enables us to calculate the lower limit for the ejection energy and the upper limit for the ejection radius, as we will see in the following Section.

IV. CALCULATION OF THE EJECTION ENERGY

Since the integral in Eq.(9) can not be solved analytically some approximations must be applied. We will restrict ourselves to the linear Stark-shift which follows from Eq.(9) when the integrand and the turning point is expanded in powers of the field g. The result corresponds to the quantum mechanical one in the limit of high quantum numbers, $n \to \infty$.

The potential barrier for H_u in the case $p_\varphi \equiv 0$ is located at $u_b = \omega/\sqrt{g}$. The value of the potential at that point is $V_u(u_b) = \omega^4/(4g)$. Since the kinetic energy is zero when the potential barrier meets the total energy $A = Z$ for the first time (this is the case due to adiabaticity), one has

$$\frac{\omega^4}{4g} = A = Z \tag{10}$$

at that certain moment.

Now, we assume a linear dependence between energy and field, i.e., only the linear Stark effect is included in our further calculations,

$$\mathcal{E}(E) = \mathcal{E}_0 + \frac{4}{\lambda}E, \qquad \lambda < 0. \tag{11}$$

As derived by expanding the invariant (9) or from the classical limit of the well-known quantum mechanical result [24]

$$\lambda = \frac{16\mathcal{E}_0}{3Z}$$

holds, which will be used in the following.

The "over the top"-criterion (10) then reads

$$\frac{\mathcal{E}^2}{\lambda(\mathcal{E} - \mathcal{E}_0)} = Z,$$

and this leads to the "over the top"-field strength

$$E_b = E(\mathcal{E}_b) = \frac{4}{9}\frac{\mathcal{E}_0^2}{Z}. \tag{12}$$

The electron is not yet free when it flows over the barrier because its Coulomb energy $\mathcal{E} - Ez$ is still negative. Demanding $\mathcal{E} - Ez = 0$, which may be written in u and v as $\mathcal{E} - g(v^2 - u^2) = 0$, is equivalent to claim

$$V_u(u) = \frac{\omega^2}{2}u^2 - \frac{1}{4}gu^4 = 0$$

when $v = 0$, as it is in our case. Therefore $p_u^2/2 = A = Z$ holds during the moment when the electron becomes free. The coordinate u_0 where this happens is easily determined through $u_0^2/4 = \omega^2/(2g)$ This equation gives an *upper* limit for the ejection radius,

$$r_0 = 3\frac{Z}{|\mathcal{E}_0|}. \tag{13}$$

The physical kinetic energy T is related to p_u according Eq.(4) (using $p_v = v = p_\varphi = 0$) through $T := 2p_u^2/u^2$. For u_0 this leads to $T_0 = 2Zg/\omega^2$. Going back to the physical quantities $\mathcal{E}$ and E instead of ω^2 and g this means $T_0 = -ZE/\mathcal{E}$. Assuming that $\mathcal{E}$ and E do not vary much during the short time between "flowing over the top" and "getting positive Coulomb energy", i.e. ionization, and inserting λ, we get

$$T_0 = \frac{1}{3}|\mathcal{E}_0|. \tag{14}$$

Note that the ejection energy is essentially the kinetic energy gained by the "u-particle" when it slides from the top of the potential barrier down to $V_u(u_0) = 0$. Multiplication with a factor $4/u_0^2$ leads to the physical ejection energy T_0.

We have already mentioned the two extreme type of orbits: they are both aligned along the z-axis (eccentricity $\to 1$), the early ionizing one on the negative half, the latest ionizing orbit on the positive half. For the latter one $u = 0$ and $A = 0$ holds instead of $v = 0$ and $B = 0$. Therefore, we have only to substitute $g \to -g$ in the result for the Stark-shift. Obviously, there is no potential barrier in $V_v(v)$ over which the electron could escape. Nevertheless, from the "zero Coulomb energy"-condition $V_v(v) = 0$ follows, as in the u-case. This condition can only be fulfilled through $v = 0$. Thus, $u = v = 0$ holds simultaneously, i.e., the electron ionizes over the Coulomb singularity in the limit eccentricity $\to 1$, and the kinetic energy is infinite at that point. So we conclude that there is no *upper* limit for the ejection energy.

V. NUMERICAL RESULTS

Our numerical simulations presented in the following were performed for the "classical" 1s state of atomic hydrogen, $Z = 1$, $\mathcal{E}_0 = -1/2$. The classical linear Stark effect then gives $\lambda = -8/3$. The "over the top"-values are calculated to be $\mathcal{E}_b = -2/3$, $E_b = 1/9$, and, finally, the lower limit for the ejection energy is found to be $T_0 = 1/6$. Eq.(13) gives for the ejection radius $r_0 = |z_0| = 6$, i.e., all electrons will be ejected within a sphere of radius 6 a.u..

Numerically, the exact motion of an ensemble of electrons in a Coulomb potential and an electric field can easily be determined using the so-called "classical trajectory Monte Carlo-method" (CTMC) [25]. We have performed CTMC runs and looked at each test electron when its Coulomb energy $\mathcal{E} - E(t)z$ becomes positive, i.e., when the electron would be free if the field is turned off immediately (this is our definition of ionization).

In Fig.3 the kinetic ejection energy of each electron is indicated by a symbol over the field strength where ionization takes place (the field is directly proportional to the time due to linear ramping) for $Z = 1$, $\mathcal{E}_0 = -1/2$, $E(t) = t/2000$, and an ensemble consisting of 1000 test electrons. The lower limit of the ejection energy is indicated in the plot by a horizontal line.

In Fig.4 a similar plot for the ejection radius is presented.

An oscillating field $E(t) \sim \cos \Omega t$ can be treated adiabatically when the frequency Ω is small compared to the orbital frequency $|\mathcal{E}_0|/\hbar$, i.e. in atomic units $\Omega/|\mathcal{E}_0| \ll 1$. We repeated our numerical run discussed in the previous paragraph with the field replaced by $E(t) = t\cos(\Omega t)/2000$, $\Omega = 0.04$. The frequency Ω corresponds to Nd laser-light and due to $\Omega/|\mathcal{E}_0| = 0.08$ an almost adiabatic behavior is expected. In Fig.5 the kinetic ejection energies of each test particle is plotted vs. its ionization time. The absolute value of the electric field and the calculated lower limit for the ejection energy are included. Apart from three test particles which are released extremely out of phase, i.e., when the electric field is low, all electrons lie above the calculated lower limit.

VI. QUANTUM MECHANICAL CALCULATIONS

The anharmonic oscillator method provides also an elegant and alternative way to derive the anomalous critical field [26] for hydrogen-like ions. With the ansatz $\psi(u, v, \varphi, \varphi'(\varphi)) = \psi_u(u)\exp(\mathrm{i}m\varphi)\psi_v(v)\exp(\mathrm{i}m'\varphi')$ the Hamiltonians (5) and (6) lead to the following two Schrödinger equations

$$\left\{-\frac{1}{2}\left[\frac{1}{u}\partial_u(u\partial_u) - \frac{m}{u^2}\right] + \frac{\omega^2}{2}u^2 - \frac{1}{4}gu^4\right\}\psi_u(u) = A\psi_u(u),$$

$$\left\{-\frac{1}{2}\left[\frac{1}{v}\partial_v(v\partial_v) - \frac{m'}{v^2}\right] + \frac{\omega^2}{2}v^2 + \frac{1}{4}gv^4\right\}\psi_v(v) = B\psi_v(v),$$

$$m = m',$$

$$A + B = Z.$$

Since the solution of the unperturbed problem, which is a 2D harmonic oscillator, is a Gaussian in the $m = m' = 0$-case, we use

$$\psi_u(u) = \sqrt{a_u/\pi}\exp(-a_u u^2/2)$$

as a trial function. The total energy then is

$$A(g) = 2\pi \int_0^\infty \mathrm{d}u\, u\psi_u^* H_u \psi_u = \frac{1}{a_u}\left(\frac{\omega^2}{2} - \frac{a_u^2}{2}\right) + a_u - \frac{g}{2a_u^2}.$$

Minimizing this energy yields up to first order in g $a_u = \omega(1 - g/\omega^3)$ and $a_v = \omega(1 + g/\omega^3)$. The oscillator energies are $A(g) = \omega(1 - g/(2\omega^3))$ and $B(g) = \omega(1 + g/(2\omega^3))$. Note that this is consistent with the fact that the linear Stark-effect vanishes for the ground state of hydrogen-like ions, since $A(g) + B(g) = 2\omega$ and from $A + B = Z$ and $\omega^2 = -\mathcal{E}_0/2$ follows $\mathcal{E}_0 = -Z^2/2$. For the hydrogen 1s-state exposed to an adiabatically ramped dc field E, the wave function in physical coordinates is

$$\psi_H(\rho, z) = \frac{1 - 4E^2}{\sqrt{\pi}} \exp(-\sqrt{\rho^2 + z^2}) \exp(-2Ez), \tag{15}$$

i.e., the unperturbed wave function is multiplied by a "deformation factor" $\exp(-2Ez)$.

In order to calculate the critical field $E_{\mathrm{crit}} = 4g_{\mathrm{crit}}$ we claim that the u-Gaussian touches the potential barrier energetically:

$$A(g) = \omega - \frac{g}{2\omega^2} = \frac{\omega^4}{4g}.$$

This leads to

$$g_{\mathrm{crit}} = \omega^3(1 - 1/\sqrt{2}) \approx 0.3\omega^3.$$

For the 1s hydrogen state we get $E^H_{\mathrm{crit}} = 0.15$ a.u. in accordance with the result presented in [26].

In Fig. 6 a comparison is made between the calculated deformation $|\psi_H(\rho, z)|^2$ which results from Eq. 15 and the exact numerical solution of the time-dependent Schrödinger equation where we ramped the field linearly over 30 atomic time units up to the critical field $E^H_{\mathrm{crit}} = 0.15$ a.u.. Strong deviation occurs only for the 1%-contour while the main parts of the probability densities agree well.

In Fig. 7 the initial probability density and the variationally determined one at the critical field are shown in the energy diagram. Note that at the critical field the packet lies 0.27 atomic energy units above the barrier in z-direction. This value may be interpreted as a quantum mechanical ejection energy.

VII. CONCLUSIONS

We have shown that within the framework of the anharmonic oscillator model important features in strong field ionization can be derived. Using this method, we have calculated (i) the lower limit for the ejection energy, $T_0 = |\mathcal{E}_0|/3$, (ii) the upper limit for the ejection radius, $r_0 = 3Z/|\mathcal{E}_0|$, and (iii) the anomalous critical field for hydrogen-like ions as well as the shape of the deformed probability density. We have also demonstrated that there exists no upper limit for the ejection energy. All results have been confirmed by CTMC simulations.

FIG. 1.

The two 2D anharmonic oscillators in the case $p_\varphi = p_{\varphi'} = 0$. Initially, when $g = 0$ holds the two potentials are pure harmonic ones (dashed). As g is adiabatically raised one expects an adiabatic lowering of level A and a raising of level B.

FIG. 2.

For the early ionizing Kepler orbit discussed in the text $v \equiv 0$ holds. Therefore, and owing to the constraint $A + B = Z$, both levels do not move when g is adiabatically raised.

FIG. 3.

The ejection energy of all test electrons in z- and ρ direction ($\diamond, \triangle$), and the sum of both (■). The calculated lower limit for the ejection energy is also shown. One can see that this limit is confirmed by the numerical runs: none of the kinetic energies ■ lies beneath the limit.

FIG. 4.

The radii of all test electron when they become free. The analytically calculated upper limit is confirmed by the numerical run. All electrons are ionized within a sphere of radius 6 atomic units.

FIG. 5.

The kinetic ejection energies in an oscillatory E-field. Only 3 of 1000 test electrons ionize through a "non-adiabatic channel" in such a way that they lie beneath the calculated lower limit. Note, that these particles become free when the field is low. The absolute value of the electric field is also shown.

FIG. 6.

The contour lines representing 80, 50, 10 and 1% of the peak height at the origin $\rho = z = 0$ are shown for the numerical result (solid) and the variationally calculated probability density (dotted). The field strength is the critical one $E = 0.15$ a.u..

FIG. 7.

Unperturbed and Stark-deformed 1s peak for atomic hydrogen at the critical field strength $E_{\rm crit} = 0.15$ a.u.. The energy gap of 0.27 a.u. between the energy level at $\mathcal{E}_0 = -1/2$ and the barrier can be interpreted as an quantum mechanical ejection energy.

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* http://www.physik.th-darmstadt.de/tqe/

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